

28th Annual Report 2019

**Convention on Long-range
Transboundary Air Pollution**

**International Cooperative Programme
on Integrated Monitoring of Air Pollution
Effects on Ecosystems**

Sirpa Kleemola and Martin Forsius (eds.)



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wge Working Group on Effects of the
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ABSTRACT

The Integrated Monitoring Programme (ICP IM) is part of the effect-oriented activities under the 1979 Convention on Long-range Transboundary Air Pollution, which covers the region of the United Nations Economic Commission for Europe (UNECE). The main aim of ICP IM is to provide a framework to observe and understand the complex changes occurring in natural/semi natural ecosystems.

This report summarizes the work carried out by the ICP IM Programme Centre and several collaborating institutes. The emphasis of the report is in the work done during the programme year 2018/2019 including:

- A short summary of previous data assessments
- A status report of the ICP IM activities, content of the IM database, and geographical coverage of the monitoring network
- An interim report on aluminium fractions in surface waters draining catchments of ICP Integrated Monitoring network
- National Reports on ICP IM activities are presented as annexes.

Keywords:

Integrated Monitoring, ecosystems, small catchments, air pollution

TIIVISTELMÄ

Ympäristön yhdennetyn seurannan ohjelma (ICP IM) kuuluu kansainvälisen ilman epäpuhtauksien kaukokulkeutumista koskevan yleissopimuksen "Convention on Long-range Transboundary Air Pollution" (1979) alaisiin seurantaohjelmiin. Yhdennetyn seurannan ohjelmassa selvitetään kaukokulkeutuvien saasteiden ja muiden ympäristömuutosten vaikutuksia elinympäristöömme. Muutosten seuranta ja ennusteita muutosten laajuudesta ja nopeudesta tehdään yleensä pienillä metsäisillä valuma-alueilla, mutta verkostoon kuuluu myös muita alueita.

Tämä julkaisu on kooste ohjelmakeskuksen ja yhteistyölaitosten toiminnasta kaudella 2018/2019, joka sisältää:

- Lyhyen yhteenvedon ohjelmassa aiemmin tehdyistä arvioinneista
- Kuvauksen ICP IM ohjelman toiminnasta ja ohjelman seurantaverkosta
- Väli raportin alumiinifraktioiden osuuksista ICP IM alueiden pintavesissä
- Kuvauksia kansallisesta ICP IM toiminnasta eri maissa liitteenä.

Asiasanat:

Yhdennetty ympäristön seuranta, ekosysteemit, pienet valuma-alueet, ilmansaasteet

SAMMANDRAG

Programmet för Integrerad övervakning av miljötillståndet (ICP IM) är en del av monitoringstrategin under UNECE:s luftvårdskonvention (LRTAP). Syftet med ICP IM är att utvärdera komplexa miljöförändringar på avrinningsområden.

Rapporten sammanfattar de utvärderingar som gjorts av ICP IM Programme Centre och de samarbetande instituten under programåret 2018/2019. Rapporten innehåller:

- En sammanfattning av programmets nuvarande omfattning och databasens innehåll
- En syntes av tidigare utvärderingar av data från programmet
- En preliminär rapport om koncentrationerna av aluminiumfraktioner i ytvatten från ICP IM områden
- Beskrivning av nationella ICP IM aktiviteter.

Nyckelord:

Integrerad miljöövervakning, ekosystem, små avrinningsområden, luftföroreningar

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ABBREVIATIONS

AMAP	Arctic Monitoring and Assessment Programme
ANC	Acid neutralising capacity
CCE	Coordination Center for Effects
CL	Critical Load
CNTER	Carbon-nitrogen interactions in forest ecosystems
ECE	Economic Commission for Europe
eLTER	The Horizon 2020 project “eLTER” (European Long-Term Ecosystem and socio-ecological Research Infrastructure) A project involving many LTER-Europe partners and sites in collaboration with the European Critical Zone Observatory community
EMEP	Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
EU	European Union
EU LIFE	EU’s financial instrument supporting environmental and nature conservation projects throughout the EU
Horizon 2020	H2020, EU Research and Innovation programme
ICP	International Cooperative Programme
ICP Forests	International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests
ICP IM	International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems
ICP Materials	International Cooperative Programme on Effects on Materials
ICP M&M	ICP Modelling and Mapping, International Cooperative Programme on Modelling and Mapping of Critical Loads and Levels and Air Pollution Effects, Risks and Trends
ICP Waters	International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes
ICP Vegetation	International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops
ILTER	International Long Term Ecological Research Network
IM	Integrated Monitoring
JEG	JEG DM, Joint Expert Group on Dynamic Modelling
LRTAP Convention	Convention on Long-range Transboundary Air Pollution
LTER-Europe	European Long-Term Ecosystem Research Network
LTER-Network	Long Term Ecological Research Network
NFP	National Focal Point
TF	Task Force
Task Force on Health	The Joint Task Force on the Health Aspects of Air Pollution
UNECE	United Nations Economic Commission for Europe
WGE	Working Group on Effects

Summary

Background and objectives of ICP IM

Integrated monitoring of ecosystems means physical, chemical and biological measurements over time of different ecosystem compartments simultaneously at the same location. In practice, monitoring is divided into a number of compartmental sub-programmes which are linked by the use of the same parameters (cross-media flux approach) and/or same or close stations (cause-effect approach).

The International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM, www.syke.fi/nature/icpim) is part of the Effects Monitoring Strategy under the Convention on Long-range Transboundary Air Pollution (LRTAP Convention). The main objectives of the ICP IM are:

- To monitor the biological, chemical and physical state of ecosystems (catchments/plots) over time in order to provide an explanation of changes in terms of causative environmental factors, including natural changes, air pollution and climate change, with the aim to provide a scientific basis for emission control.
- To develop and validate models for the simulation of ecosystem responses and use them (a) to estimate responses to actual or predicted changes in pollution stress, and (b) in concert with survey data to make regional assessments.
- To carry out biomonitoring to detect natural changes, in particular to assess effects of air pollutants and climate change.

The full implementation of the ICP IM will allow ecological effects of heavy metals, persistent organic substances and tropospheric ozone to be determined. A primary concern is the provision of scientific and statistically reliable data that can be used in modelling and decision making.

The ICP IM sites (mostly forested catchments) are located in undisturbed areas, such as natural parks or comparable areas. The ICP IM network presently covers forty-nine sites from sixteen countries. The international Programme Centre is located at the Finnish Environment Institute in Helsinki. The present status of the monitoring activities is described in detail in Chapter 1 of this report.

A manual detailing the protocols for monitoring each of the necessary physical, chemical and biological parameters is applied throughout the programme (Manual for Integrated Monitoring 1998, and updated web version).

Assessment activities within the ICP IM

Assessment of data collected in the ICP IM framework is carried out at both national and international levels. Key tasks regarding international ICP IM data have been:

- Input-output and proton budgets
- Trend analysis of bulk and throughfall deposition and runoff water chemistry
- Assessment of responses using biological data
- Dynamic modelling and assessment of the effects of different emission / deposition scenarios, including confounding effects of climate change processes
- Assessment of concentrations, pools and fluxes of heavy metals
- Calculation of critical loads for sulphur and nitrogen compounds, and assessment of critical load exceedance, as well as links between critical load exceedance and empirical impact indicators.

Conclusions from international studies using ICP IM data

Input-output and proton budgets, C/N interactions

Ion mass budgets have proved to be useful for evaluating the importance of various biogeochemical processes that regulate the buffering properties in ecosystems. Long-term monitoring of mass balances and ion ratios in catchments/plots can also serve as an early warning system to identify the ecological effects of different anthropogenically derived pollutants, and to verify the effects of emission reductions.

The most recent results from ICP IM studies are available from the study of Vuorenmaa et al. (2017). Site-specific annual input-output budgets were calculated for sulphate (SO_4) and total inorganic nitrogen ($\text{TIN} = \text{NO}_3\text{-N} + \text{NH}_4\text{-N}$) for 17 European ICP IM sites in 1990–2012. Temporal trends for input (deposition) and output (runoff water) fluxes and net retention/net release of SO_4 and TIN were also analysed. Large spatial variability in the input and output fluxes of SO_4 and TIN reflects important gradients of air pollution effects in Europe, with the highest deposition and runoff water fluxes in southern Scandinavia, Central and Eastern Europe and the lowest fluxes at more remote sites in northern European regions. A significant decrease in the total (wet + dry) non-marine SO_4 deposition and bulk deposition of TIN was found at 90% and 65% of the sites, respectively. Output fluxes of non-marine SO_4 in runoff decreased significantly at 65% of the sites, indicating positive effects of international emission abatement actions in Europe during the last 25 years. Catchments retained SO_4 in the early and mid-1990s, but this shifted towards a net release in the late 1990s, which may be due to the mobilization of legacy S pools accumulated during times of high atmospheric SO_4 deposition. Despite decreased deposition, TIN output fluxes and retention rates showed a mixed response with both decreasing (9 sites) and increasing (8 sites) trend slopes, but trends were rarely significant. In general, TIN was strongly retained in the catchments not affected by natural disturbances. The long-term annual variation in net releases for SO_4 was explained by variations in runoff and SO_4 concentrations in deposition, while a variation in TIN concentrations in runoff was mostly associated with a variation of the TIN retention rate in catchments. Net losses of SO_4 may lead to a slower recovery of surface waters than those predicted by the decrease in SO_4 deposition. Continued enrichment of N in catchment soils poses a threat to terrestrial biodiversity and may ultimately lead to higher TIN runoff through N saturation or climate change. Continued monitoring and further evaluations of mass balance budgets are thus needed.

Earlier results from ICP IM studies are summarized below.

The first results of input-output and proton budget calculations were presented in the 4th Annual Synoptic Report (ICP IM Programme Centre 1995) and the updated results regarding the effects of N deposition were presented in Forsius et al. (1996). Data from selected ICP IM sites were also included in European studies for evaluating soil organic horizon C/N-ratio as an indicator of nitrate leaching (Dise et al. 1998, MacDonald et al. 2002). Results regarding the calculation of fluxes and trends of S and N compounds were presented in a scientific paper prepared for the Acid Rain Conference, Japan, December 2000 (Forsius et al. 2001). A scientific paper regarding calculations of proton budgets was published in 2005 (Forsius et al. 2005).

The budget calculations showed that there was a large difference between the sites regarding the relative importance of the various processes involved in the transfer of acidity. These differences reflected both the gradients in deposition inputs and

the differences in site characteristics. The proton budget calculations showed a clear relationship between the net acidifying effect of nitrogen processes and the amount of N deposition. When the deposition increases also N processes become increasingly important as net sources of acidity.

A critical deposition threshold of about 8–10 kg N ha⁻¹ yr⁻¹, indicated by several previous assessments, was confirmed by the input-output calculations with the ICP IM data (Forsius et al. 2001). The output flux of nitrogen was strongly correlated with key ecosystem variables like N deposition, N concentration in organic matter and current year needles, and N flux in litterfall (Forsius et al. 1996). Soil organic horizon C/N-ratio seems to give a reasonable estimate of the annual export flux of N for European forested sites receiving throughfall deposition of N up to about 30 kg N ha⁻¹ yr⁻¹. When stratifying data based on C/N ratios less than or equal to 25 and greater than 25, highly significant relationships were observed between N input and nitrate leached (Dise et al. 1998, MacDonald et al. 2002, Gundersen et al. 2006). Such statistical relationships from intensively studied sites can be efficiently used in conjugation with regional monitoring data (e.g. ICP Forests and ICP Waters data) in order to link process level data with regional-scale questions.

An assessment on changes in the retention and release of S and N compounds at the ICP IM sites was prepared for the 21st Annual Report (Vuorenmaa et al. 2012). Updated and revised data were included in the continuation of the work in the 22nd and 23rd Annual Reports (Vuorenmaa et al. 2013, 2014). The relationship between N deposition and organic N loss and the role of organic nitrogen in the total nitrogen output fluxes were derived in Vuorenmaa et al. (2013).

Sulphur budgets calculations indicated a net release of S from many ICP IM sites, indicating that the soils are releasing previously accumulated S. Similar results have been obtained in other European plot and catchment studies.

The reduction in deposition of S and N compounds at the ICP IM sites, caused by the “Protocol to Abate Acidification, Eutrophication and Ground-level Ozone” of the LRTAP Convention (“Gothenburg protocol”), was estimated for the year 2010 using transfer matrices and official emissions. Implementation of the protocol will further decrease the deposition of S and N at the ICP IM sites in western and north western parts of Europe, but in more eastern parts the decrease will be smaller (Forsius et al. 2001).

Results from the ICP IM sites were also summarised in an assessment report prepared by the Working Group on Effects of the LRTAP Convention (WGE) (Sliggers & Kakebeeke 2004, Working Group on Effects 2004).

ICP IM contributed to an assessment report on reactive nitrogen (N_r) of the WGE. This report was prepared for submission to the TF on Reactive Nitrogen and other bodies of the LRTAP Convention to show what relevant information has been collected by the ICP programmes under the aegis of the WGE to allow a better understanding of N_r effects in the ECE region. The report contributed relevant information for the revision of the Gothenburg Protocol. A revised Gothenburg Protocol was successfully finalised in 2012.

It should also be recognized that there are important links between N deposition and the sequestration of C in the ecosystems (and thus direct links to climate change processes). These questions were studied in the CNTER-project in which data from both the ICP IM and EU/Intensive Monitoring sites were used (Gundersen et al. 2006). A summary report of the CNTER-results on C/N -interactions and nitrogen effects in European forest ecosystems was prepared for the WGE meeting 2007 (ECE/EB.AIR/WG.1/2007/10).

Trend analysis

Empirical evidence on the development of environmental effects is of central importance for the assessment of success of international emission reduction policy. In order to assess the impacts of air pollution and climate change in the environment, a long-term integrated monitoring approach in remote unmanaged areas including physical, chemical and biological variables is needed. Vuorenmaa et al. (2018) evaluated long-term trends (1990–2015) for deposition and runoff water chemistry and fluxes, and climatic variables at 25 ICP IM sites in Europe that commonly belong also to the LTER-Europe/ILTER networks. The trend assessment was published in a special issue in *Science of the Total Environment* with the title: “International Long-Term Ecological Research (ILTER) network”. The recent results from trend assessment at IM sites confirm that emission abatement actions are having their intended effects on precipitation and runoff water chemistry in the course of successful emission reductions in different regions in Europe. Concentrations and deposition fluxes of xSO_4 , and consequently acidity in precipitation, have substantially decreased in IM areas. Inorganic N (TIN) deposition has decreased in most of the IM areas, but to a lesser extent than that of xSO_4 . Substantially decreased xSO_4 deposition has resulted in decreased concentrations and output fluxes of xSO_4 in runoff, and decreasing trends of TIN concentrations in runoff – particularly for NO_3 – are more prominent than increasing trends. In addition, decreasing trends appeared to strengthen over the course of emission reductions during the last 25 years. TIN concentrations in runoff were mainly decreasing, while trends in output fluxes were more variable, but trend slopes were decreasing rather than increasing. However, decreasing trends for S and N emissions and deposition and deposition reduction responses in runoff water chemistry tended to be more gradual since the early 2000s. Air temperature increased significantly at 61% of the sites, while trends for precipitation and runoff were rarely significant. The site-specific variation of xSO_4 concentrations in runoff was most strongly explained by deposition. Climatic variables and deposition explained the variation of TIN concentrations in runoff at single sites poorly, and as yet there are no clear signs of a consistent deposition-driven or climate-driven increase in TIN exports in the catchments.

Vuorenmaa et al. (2018) reported that the IM sites are located in areas with very different N deposition gradients, and it is obvious that not all potential drivers were included in the empirical model in the study, and further analysis with specific landscape and soil data is needed to elucidate the variation in inorganic N concentrations in runoff at IM sites. Thus, the next phase of the work on trend assessment will be an assessment of the role of internal nitrogen parameters (Vuorenmaa et al. in prep.).

Earlier work is summarized below.

First results from a trend analysis of monthly ICP IM data on bulk and throughfall deposition as well as runoff water chemistry were presented in Vuorenmaa (1997). ICP IM data on water chemistry were also used for a trend analysis carried out by the ICP Waters and results were presented in the Nine Year Report of that programme (Lükewille et al. 1997).

Calculations on the trends of N and S compounds, base cations and hydrogen ions were made for 22 ICP IM sites with available data across Europe (Forsius et al. 2001). The site-specific trends were calculated for deposition and runoff water fluxes using monthly data and non-parametric methods. Statistically significant downward trends of SO_4 , NO_3 and NH_4 bulk deposition (fluxes or concentrations) were observed at 50% of the ICP IM sites. Sites with higher N deposition and lower C/N-ratios clearly showed higher N output fluxes, and the results were consistent with previous obser-

variations from European forested ecosystems. Decreasing SO_4 and base cation trends in runoff waters were commonly observed at the ICP IM sites. At some sites in the Nordic countries decreasing NO_3 and H^+ trends (increasing pH) were also observed. The results partly confirmed the effective implementation of emission reduction policy in Europe. However, clear responses were not observed at all sites, showing that recovery at many sensitive sites can be slow and that the response at individual sites may vary greatly.

Data from ICP IM sites were also used in a study of the long-term changes and recovery at nine calibrated catchments in Norway, Sweden and Finland (Moldan et al. 2001, RECOVER: 2010 project). Runoff responses to the decreasing deposition trends were rapid and clear at the nine catchments. Trends at all catchments showed the same general picture as from small lakes in Scandinavia.

It was agreed at the ICP IM Task Force meeting in 2004 that a new trend analysis should be carried out. The preliminary results were presented in Kleemola (2005) and the updated results in the 15th Annual Report (Kleemola & Forsius 2006). Statistically significant decreases in SO_4 concentrations were observed at a majority of sites in both deposition and runoff/soil water quality. Increases in ANC (acid neutralising capacity) were also commonly observed. For NO_3 the situation was more complex, with fewer decreasing trends in deposition and even some increasing trends in runoff/soil water.

Results from several ICPs and EMEP were used in an assessment report on acidifying pollutants, arctic haze and acidification in the arctic region prepared for the Arctic Monitoring and Assessment Programme (AMAP, Forsius & Nyman 2006, www.amap.no). Sulphate concentrations in air showed generally decreasing trends since the 1990s. In contrast, levels of nitrate aerosol were increasing during the arctic haze season at two stations in the Canadian arctic and Alaska, indicating a decoupling between the trends in sulphur and nitrogen. Chemical monitoring data showed that lakes in the Euro-Arctic Barents region are showing regional scale recovery. Direct effects of sulphur dioxide emissions on trees, dwarf shrubs and epiphytic lichens were observed close to large smelter point sources.

The recent trend assessment using monthly ICP IM data (Vuorenmaa et al. 2018) was preceded by corresponding trend evaluations for the periods 1993–2006 and 1990–2013 (Vuorenmaa et al. 2009, 2016, respectively). Moreover, trends for annual input and output fluxes of SO_4 and TIN were evaluated for the period 1990–2012 (Vuorenmaa et al. 2017). These results clearly showed the regional-scale decreasing trends of SO_4 in deposition and runoff/soil water, and suggested that IM catchments have increasingly responded to the decreases in S emissions and depositions of SO_4 since the early 1990s. Decreased nitrogen emissions also resulted in decrease of inorganic N deposition, but to a lesser extent than that of SO_4 , and trends in TIN fluxes in runoff were highly variable due to complex processes in terrestrial catchment that are not yet fully understood. Besides, the net release of SO_4 in forested catchments fueled by the mobilization of legacy S pools, accumulated during times of high atmospheric sulphur deposition, may delay the recovery from acidification. The more efficient retention of inorganic N than SO_4 results in generally higher leaching fluxes of SO_4 than those of inorganic N in European forested ecosystems. SO_4 thus remains the dominant source of actual soil acidification despite the generally lower input of SO_4 than inorganic N. Critical load calculations for Europe also indicated exceedances of the N critical loads over large areas. Long-term trends for deposition and runoff variables were for the first time evaluated together with climatic variables (precipitation, runoff water volume and air temperature) at IM sites by Vuorenmaa et al. (2016). Many study sites exhibited long-term seasonal trends with a significant increase in air temperature, precipitation and runoff particularly in spring and autumn, but annual trends were rarely significant. It was concluded that the sulphur

and nitrogen problem thus clearly requires continued attention as a European air pollution issue, and further long-term monitoring and trend assessments of different ecosystem compartments and climatic variables are needed to evaluate the effects, not only of emission reduction policies, but also of changing climate.

An assessment on changes in the retention and release of S and N compounds at the ICP IM sites was prepared for the 21st Annual Report (Vuorenmaa et al. 2012). Updated and revised data were included in the continuation of the work in the 22nd and 23rd Annual Reports. The role of organic nitrogen in mass balance budget was derived and trends of S and N in fluxes were analysed (Vuorenmaa et al. 2013, 2014).

Detected responses in biological data

The effect of pollutant deposition on natural vegetation, including both trees and understorey vegetation, is one of the central concerns in the impact assessment and prediction. The most recent ICP IM study on dose-response relationships was published by Dirnböck et al. (2014). This study utilized a new ICP IM database for biological data and focussed on effects on forest floor vegetation from elevated nitrogen deposition. Results on dynamic modelling of vegetation responses have also recently been published (Dirnböck et al. 2018, see below)

In many European countries airborne nitrogen coming from agriculture and fossil fuel burning exceeds critical thresholds and threatens the functioning of ecosystems. One effect is that high levels of nitrogen stimulate the growth of only a few plants that outcompete other, often rare, species. As a consequence biodiversity declines. Though this is known to happen in natural and semi-natural grasslands, it has never been shown in forest ecosystems where management is a strong, mostly overriding determinant of biodiversity. Dirnböck et al. (2014) utilized long-term monitoring data from 28 Integrated Monitoring sites to analyse temporal trends in plant species cover and diversity. At sites where nitrogen deposition exceeded the critical load, the cover of forest plant species preferring nutrient-poor soils (oligotrophic species) significantly decreased whereas plant species preferring nutrient-rich soils (eutrophic species) showed – though weak – an opposite trend. These results show that airborne nitrogen has changed the structure and composition of forest floor vegetation in Europe. Plant species diversity did not decrease significantly within the observed period but the majority of newly established species was found to be eutrophic. Hence it was hypothesized that without reducing nitrogen deposition below the critical load forest biodiversity will decline in the future.

Previous work on biological data is summarized below.

The first assessment of vegetation monitoring data at ICP IM sites with regards to N and S deposition was carried out by Liu (1996). Vegetation monitoring was found useful in reflecting the effects of atmospheric deposition and soil water chemistry, especially regarding sulphur and nitrogen. The results suggested that plants respond to N deposition more directly than to S deposition with respect to vegetation indices. De Zwart (1998) carried out an exploratory multivariate statistical gradient analysis of possible causes underlying the aspect of forest damage at ICP IM sites. These results suggested that coniferous defoliation, discolouration and lifespan of needles in the diverse phenomena of forest damage are for respectively 18%, 42% and 55% explained by the combined action of ozone and acidifying sulphur and nitrogen compounds in air.

As a separate exercise, the epiphytic lichen flora of 25 European ICP IM monitoring sites, all situated in areas remote from local air pollution sources, was statistically related to measured levels of SO₂ in air, NH₄⁺, NO₃⁻ and SO₄²⁻ in precipitation, annual bulk precipitation, and annual average temperature (van Herk et al. 2003, de Zwart

et al. 2003). It was concluded that long distance transport of nitrogen air pollution is important in determining the occurrence of acidophytic lichen species, and constitutes a threat to natural populations that is strongly underestimated so far.

In 2010, the Task Force meeting decided upon a new reporting format for biological data. The new format was based on primary raw data, and not aggregated mean values as before. All countries were encouraged to re-report old data in the new format. This was successful and as a result, the full potential of the biological data from the ICP Integrated Monitoring network could be utilised to raise and answer research question that the old database could not.

Dynamic modelling and assessment of the effects of emission/deposition scenarios

In a policy-oriented framework, dynamic models are needed to explore the temporal aspect of ecosystem protection and recovery. The critical load concept, used for defining the environmental protection levels, does not reveal the time scales of recovery. Priority in the ICP IM work is given to site-specific modelling. The role of ICP IM is to provide detailed and consistent physical and chemical data and long time-series of observations for key sites against which model performance can be assessed and key uncertainties identified (see Jenkins et al. 2003). ICP IM participates also in the work of the Joint Expert Group on Dynamic Modelling (JEG) of the WGE.

Dynamic vegetation modelling at ICP IM sites has been conducted with contributions from ICP M&M, ICP Forests, and the LTER Europe network. The VSD+ model was applied to simulate soil chemistry at 26 sites in ten countries throughout Europe (Holmberg & Dirnböck 2015, 2016, Dirnböck et al. 2018a; 2018b, Holmberg et al. 2018). Simulated future soil conditions improved under projected decrease in deposition and current climate conditions: higher pH, BS and C:N at 21, 16 and 12 of the sites, respectively. Dirnböck et al. (2018b) found, however, that a release from eutrophication is not expected to result from the decrease in N deposition under current legislation emission (CLE) reduction targets until 2030.

Dynamic models have also previously been developed and used for the emission/deposition and climate change scenario assessment at several selected ICP IM sites (e.g. Forsius et al. 1997, 1998a, 1998b, Posch et al. 1997, Jenkins et al. 2003, Futter et al. 2008, 2009). These models are flexible and can be adjusted for the assessment of alternative scenarios of policy importance. The modelling studies have shown that the recovery of soil and water quality of the ecosystems is determined by both the amount and the time of implementation of emission reductions. According to the models, the timing of emission reductions determines the state of recovery over a short time scale (up to 30 years). The quicker the target level of reductions is achieved, the more rapidly the surface water and soil status recover. For the long-term response (> 30 years), the magnitude of emission reductions is more important than the timing of the reduction. The model simulations also indicate that N emission controls are very important to enable the maximum recovery in response to S emission reductions. Increased nitrogen leaching has the potential to not only offset the recovery predicted in response to S emission reduction, but further to promote substantial deterioration in pH status of freshwaters and other N pollution problems in some areas of Europe.

Work has also been conducted to predict potential climate change impacts on air pollution related processes at the sites. The large EU-project Euro-limpacs (2004–2009) studied the global change impacts on freshwater ecosystems. The institutes involved in the project used data collected at ICP IM and ICP Waters sites as key datasets for the modelling, time-series and experimental work of the project. A modelling assessment on the global change impacts on acidification recovery was carried out in the project (Wright et al. 2006). The results showed that climate/global change induced changes

may clearly have a large impact on future acidification recovery patterns, and need to be addressed if reliable future predictions are wanted (decadal time scale). However, the relative significance of the different scenarios was to a large extent determined by site-specific characteristics. For example, changes in sea-salt deposition were only important at coastal sites and changes in decomposition of organic matter at sites which are already nitrogen saturated.

In response to environmental concerns, the use of biomass energy has become an important mitigation strategy against climate change. A summary report on links between climate change and air pollution effects, based on results of the Euro-limpacs project, was prepared for the WGE meeting 2008 (ECE/EB.AIR/WG.1/2008/10). It was concluded that the increased use of forest harvest residues for biofuel production is predicted to have a significant negative influence on the base cation budgets causing re-acidification at the study catchments. Sustainable forestry management policies would need to consider the combined impact of air pollution and harvesting practices.

Pools and fluxes of heavy metals

The work to assess concentrations, stores and fluxes of heavy metals at ICP IM sites is led by Sweden. In 26th Annual Report data on Pb, Cd, Hg, Cu and Zn from countries in the ICP IM were presented (Åkerblom & Lundin 2017). These data will be used for establishment of background heavy metal concentrations in forested compartments and risk assessments of heavy metals. In many national studies on ICP IM sites, detailed site-specific budget calculations of heavy metals (including Hg) have improved the scientific understanding of ecosystem processes, retention times and critical thresholds. ICP IM sites are also used for dynamic model development of these compounds.

For the future evaluation of emission reductions of heavy metals to the atmosphere we will analyze site-specific long-term trends for fluxes of heavy metals (primarily for Cd, Pb, and Hg and depending on availability of data, also Cu and Zn) in deposition (input) and runoff (output), using available long-term monthly data collected across ICP IM sites in Europe. This will be done to see if fluxes of heavy metals in deposition and runoff respond to changes in emission reductions in Europe. Reduction in heavy metal emissions is hypothesized to be reflected in decreasing heavy metal concentrations (Åkerblom & Lundin 2015), taking into account climatic variation over time and between regions also in decreasing heavy metal fluxes. Temporal trend analysis in heavy metal fluxes will provide a detailed understanding of responses in heavy metal mass balances to emission reductions and give indication on possible change in retention of heavy metals in catchments over time. This overview will also provide an estimate on the significance in heavy metal mass balances over time and identify uncertainties in the mass balances and needs for improvements. The aim is to present results in an international scientific journal.

Increases or no change in Hg concentrations in the upper-most forest soil mor-layer does not correspond to the general decrease in emissions of Hg to the atmosphere and atmospheric concentrations (Åkerblom & Lundin 2015). Apparently there is an insufficient understanding of the governing processes and a need for more detailed data on the Hg cycling in forest catchments. One process that is not accounted for in ICP IM programme is the land-atmosphere exchange of Hg. The phenomena of land-atmosphere exchange has been known for long time but it has been quantified only recently due to the development of micrometeorological systems for continuous measurements (Osterwalder et al. 2016). In the case of mass balance calculations for Hg new evidence has shown that land-atmosphere exchange during a 2-year study over a peatland can be more than double the flux in stream runoff (Osterwalder et al. 2017). Based on natural Hg stable isotope studies in podzols and histosols, significant

Hg re-emission from organic soil horizons occurred (Jiskra et al. 2015). These novel observations and knowledge about processes that govern land-atmosphere exchange of Hg calls for methods and approaches to account for this important flux in the catchment cycle of Hg within ICP IM.

Previous work on heavy metals is summarized below.

Preliminary results on concentrations, fluxes and catchment retention were reported to the Working Group on Effects in 2001 (document EB.AIR/WG.1/2001/10). The main findings on heavy metals budgets and critical loads at ICP IM sites were presented by Bringmark (2011). Input/output budgets and catchment retention for Cd, Pb and Hg in the years 1997–2011 were determined for 14 ICP IM catchments across Europe (Bringmark et al. 2013). Litterfall plus throughfall was taken as a measure of the total deposition of Pb and Hg (wet + dry) on the basis of evidence suggesting that, for these metals, internal circulation is negligible. The same is not true for Cd. Excluding a few sites with high discharge, between 74 and 94 % of the input, Pb was retained within the catchments; significant Cd retention was also observed. High losses of Pb ($>1.4 \text{ mg m}^{-2} \text{ yr}^{-1}$) and Cd ($>0.15 \text{ mg m}^{-2} \text{ yr}^{-1}$) were observed in two mountainous Central European sites with high water discharge. All other sites had outputs below or equal to 0.36 and $0.06 \text{ mg m}^{-2} \text{ yr}^{-1}$, respectively, for the two metals. Almost complete retention of Hg, 86–99 % of input, was reported in the Swedish sites. These high levels of metal retention were maintained even in the face of recent dramatic reductions in pollutant loads. In the Progress report on heavy metal trends at ICP IM sites (Åkerblom & Lundin 2015) temporal trends were seen in forest floor with decreasing concentrations for Cd and Pb while Hg did not change. An increase in heavy metal concentrations was also seen in deeper mineral soil horizon indicating a translocation of heavy metals from upper to deeper soil horizons.

Calculation of critical loads and their exceedance, relationships to effect indicators

Empirical impact indicators of acidification and eutrophication were determined from stream water chemistry and runoff observations at ICP IM catchments (Holmberg et al. 2013). The indicators were compared with exceedances of critical loads of acidification and eutrophication obtained with deposition estimates for the year 2000. Empirical impact indicators agreed well with the calculated exceedances. Annual mean fluxes and concentrations of acid neutralizing capacity (ANC) were negatively correlated with the exceedance of critical loads of acidification. Observed leaching of nitrogen was positively correlated with the exceedances of critical loads (Holmberg et al. 2013). This study was revisited with new data on N concentrations and fluxes (Holmberg et al. 2017). For most sites, there was an improvement visible as a shift towards less exceedance and lower concentrations of total inorganic nitrogen (TIN) in runoff. At the majority of the sites both the input and the output flux of TIN decreased between the two observation periods 2000–2002 and 2013–2015. Data from the ICP IM provide evidence of a connection between modelled critical loads and empirical monitoring results for acidification parameters and nutrient nitrogen.

Planned activities

- Maintenance and development of a central ICP IM database at the Programme Centre.
- Continued assessment of the long-term effects of air pollutants to support the implementation of emission reduction protocols, including:
 - Assessment of trends.
 - Calculation of ecosystem budgets, empirical deposition thresholds and site-specific critical loads.
 - Dynamic modelling and scenario assessment.
 - Comparison of calculated critical load exceedances with observed ecosystem effects.
- Calculation of pools and fluxes of heavy metals at selected sites.
- Assessment of cause-effect relationships for biological data, particularly vegetation.
- Coordination of work and cooperation with other ICPs, particularly regarding dynamic modelling (all ICPs), cause-effect relationships in terrestrial systems (ICP Forests, ICP Vegetation), and surface waters (ICP Waters).
- Participation in the development of the European LTER-network (Long Term Ecosystem Research Network, www.lter-europe.net) to a legal entity on the ESFRI roadmap of recognised research infrastructures.
- Cooperation with other external organisations and programmes, particularly the International Long Term Ecological Research Network (ILTER, www.ilter.network, Mirtl et al. 2018).
- Participation in projects with a global change perspective.

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1 ICP IM activities, monitoring sites and available data

1.1 Review of the ICP IM activities in 2018–2019

Meetings

- Thomas Dirnböck, Environment Agency Austria, represented ICP IM in the 33rd Task Force meeting of ICP Modelling & Mapping and Joint Expert Group on Dynamic Modelling in Bern, Switzerland, 18–20 April 2018.
- ICP IM Programme Manager Martin Forsius and Co-Chair Ulf Grandin participated in the eLTER H2020 annual meeting and development of eLTER Research Infrastructure in Sofia, Bulgaria, 23–25 May 2018.
- The former ICP IM Chair Lars Lundin represented ICP IM in the 34th Task Force Meeting of ICP Forests in Riga, Latvia, 23–25 May 2018.
- Martin Forsius and Ulf Grandin represented ICP IM and Co-Chair Salar Valinia represented Sweden in the Fourth Joint Session of the Working Group on Effects and the Steering Body to EMEP in Geneva, Switzerland, 10–14 September 2018.
- Martin Forsius participated in the joint 2018 conference organized by ILTER and its East Asia Pacific (EAP) Network in Taichung City, Taiwan, 15–19 October 2018.
- Ulf Grandin represented ICP IM and Salar Valinia represented Sweden at the NEC Directive Ecosystem Monitoring subgroup meeting in Brussels, Belgium, 30 November 2018.
- Ulf Grandin, Martin Forsius and Maria Holmberg took part in the eLTER project planning meeting for the eLTER ESFRI process in Rome, Italy, 5–6 December 2018.
- Martin Forsius participated in the ‘Global event on clean air’, a special session at the 38th session of the Executive Body to the Convention on Long-range Transboundary Air Pollution (LRTAP) in Geneva, Switzerland, 12–13 December 2018.
- Martin Forsius took part in the eLTER plus meeting in Helsinki, Finland, 21–23 January 2019.
- Ulf Grandin participated in the eLTER project planning meeting for the eLTER ESFRI process in Frankfurt, Germany, 19–20 February 2019.
- Ulf Grandin, Salar Valinia and Martin Forsius represented ICP IM in the EMEP Steering Body and Working Group on Effects Bureau meeting in Laxenburg, Austria, 19–21 March 2019.
- Ulf Grandin represented ICP IM and Salar Valinia represented Sweden at the NEC Directive Ecosystem Monitoring subgroup meeting in Brussels, Belgium, 2 April 2019.
- Martin Forsius and Ulf Grandin attended the final Annual Meeting for participants of the eLTER H2020 Starting Communities project in Dalmahoy, United Kingdom, 13–16 May 2019.

- The twenty-seventh meeting of the Programme Task Force on ICP Integrated Monitoring was organized as a joint 2019 Task Force Meeting of ICP Waters and ICP Integrated Monitoring in Helsinki, Finland from June 4 to June 6, 2019.

Projects, data issues

After December 1st 2018 the National Focal Points (NFPs) reported their 2017 results to the ICP IM Programme Centre. The Programme Centre carried out standard check-up of the results and incorporated them into the IM database.

Scientific work in priority topics

- The Programme Centre prepared the ICP IM contribution to the Joint Report 2018 of the ICPs, TF health and Joint Expert Group on Dynamic Modelling for the WGE (ECE/EB.AIR/GE.1/2018/3– ECE/EB.AIR/WG.1/2018/3).
- ICP IM participates in a joint coordinated exercise on dynamic modelling together with other ICPs (Joint Expert Group on Dynamic Modelling, JEG DM). Priority in the ICP IM work is given to site-specific modelling activities and development/testing of new methodologies for assessing the connections between air pollution and climate change.

I.2 Activities and tasks planned for 2019–2020

Activities/tasks related to the programme's present objectives, carried out in close collaboration with other ICPs/ Task Forces

According to the ICP IM work plan, ICP IM will produce the following reports/papers:

- 2019/20: Report/paper on the relationship between critical load exceedances and empirical ecosystem impact indicators
- 2020: Progress report on heavy metal trends in concentrations and fluxes across ICP IM sites in Europe, potentially in cooperation with ICP Waters.
- 2019/20: Scientific paper on the impacts of catchment characteristics, climate and hydrology on N processes
- 2020: Scientific paper on the recovery in the epiphytic lichen community in the IM catchments, after the decrease in S deposition

Other activities

- Maintenance and development of central ICP IM database at the Programme Centre
- Arrangement of the 28th Task Force meeting (2020)
- Preparation of the 29th ICP IM Annual Report (2020)
- Preparation of the ICP IM contribution to assessment reports of the WGE
- Participation in meetings of the WGE, other ICPs and the JEG DM

Activities/tasks aimed at further development of the programme

- Participation in the development of the European LTER-network (Long Term Ecosystem Research network, www.lter-europe.net) to a legal entity on the ESFRI roadmap of recognised research infrastructures
- Participation in the activities of other external organisations, particularly the International Long Term Ecological Research Network (ILTER, www.ilter.network)

I.3 Published reports and articles 2018–2019

Evaluations of international ICP IM data and related publications

- Dirnböck, T., Pröll, G., Austnes, K., Beloica, J., Beudert, B., Canullo, R., De Marco, A., Fornasier, M.F., Fütter, M., Georgen, K., Grandin, U., Holmberg, M., Lindroos, A.-J., Mirtl, M., Neiryneck, J., Pecka, T., Nieminen, T.M., Nordbakken, J.-F., Posch, M., Reinds, G.-J., Rowe, E.C., Salemaa, M., Scheuschner, T., Starlinger, F., Uziębło, A.K., Valinia, S., Weldon, J., Wamelink, W. & Forsius, M. 2018. Currently legislated decreases in nitrogen deposition will yield only limited plant species recovery in European forests. *Environmental Research Letters* 13: 125010.
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I.4 Monitoring sites and data

The following countries have continued data submission to the ICP IM database during the period 2014–2018: Austria, Belarus, the Czech Republic, Estonia, Finland, Germany, Ireland, Italy, Lithuania, Norway, Poland, the Russian Federation, Spain, Sweden, Switzerland and Ukraine. Poland rejoined the network and included several sites in 2017.

The number of sites with on-going data submission for at least part of the data years 2013–2017 is 49 from sixteen countries. Sites from Canada, Latvia and United Kingdom only contain older data.

An overview of the data reported internationally to the ICP IM database is given in Table 1.1. Additional earlier reported data are available from sites outside those presented in Table 1.1. and Fig. 1.1. Locations of the ICP IM monitoring sites are shown in Fig. 1.1

Table 1.1. Internationally reported data from ICP IM sites (- subprogramme not possible to carry out, * or forest health parameters in former Forest stands/Trees).

AREA		SUBPROGRAMME*																							
		AM	AC	PC	MC	TF	SF	SC	SW	GW	RW	LC	FC	LF	RB	LB	FD	VG	BI	VS	EP	AL	MB	BB	BV
		meteorology	air chemistry	precipitation chemistry	moss chemistry	throughfall	stemflow	soil chemistry	soil water chemistry	groundwater chemistry	runoff water chemistry	lake water chemistry	foliage chemistry	litterfall	hydrobiology of streams	hydrobiology of lakes	forest damage	vegetation	bioelements	vegetation structure	epiphytes	trunk epiphytes	aerial green algae	microbial decomposition	bird inventory
AT01	ZÖBELBODEN	95-17	95-17	93-17		93-17	99-04	04	93-17		95-17	-	92-17	93-17				93			93-98				
BY02	BEREZINA BR	89-15	89-15	89-15				95-98			95-15														
CH02	LAGO NERO	15-17	15-17	15-17					17		15-17	15-16					17								
CZ01	ANENSKÉ POVODI	89-17	89-17	89-17	89	89-17		02-15	07-17	08-17	89-17	-			07	-									
CZ02	LYSINA	67-18	93-96	90-17		91-17		93	90-17	89-17	89-17	91-17	94	08	07	11		15	94			14-15		10	
DE01	FORELLENBACH	90-17	90-17	90-17	90	90-17	90-05	90-11	90-17	88-17	90-17	-	90-17	90-17		-	90-14	90-08		00	92-95		94-17	91-02	90-95
DE02	NEUGLOBSOW	67-17	98-17	98-17		98-17	04-17	04-16	98-17	98-17		98-17	06-17	04-17			04-17								
EE01	VILSANDI	95-17	94-17	94-17	94-15	94-17	94-17	94-15	94-17	95-96	-	-	94-17	94-17	-	-	94-17				94-04		94-17		94
EE02	SAAREJÄRVE	94-17	98-17	94-17	94-16	94-17	94-17	94-15	95-17	95-14	94-17	96	94-17	94-17			96-17	96-12	12		94-15	94-17	96-17	98-14	
ES02	BERTIZ	08-16	08-16	07-16		07-16	08-16	10-15	07-16		07-16		08-16	08-16			07-12	07		07					
FI01	VALKEA-KOTINEN	88-17	94-17	88-17	88-96	89-17	89-99	88-89	89-01		88-17	87-17	88-01	90-16		90-93	88-91	88-09			88-97		90	87-89	87
FI03	HIETAJÄRVI	88-17	93-00	88-17	89-96	89-17	89-99	88	89-01		88-17	87-17	88-01	90-16		90	88-91	90-09			90-97		90-91	87-89	
FI06	PALLASJÄRVI			14-17		16-17					04-17	04-17		07-16										88-89	
IE01	BRACKLOON WOOD			91-16		91-11	92-97		91-16		-	91-96	91-98	-	-										
IT01	RENON-RITTEN	90-17	93-17	93-14		93-13	93-13	93-11	93-13		00-13	-	93-10	00	-	-	92-13	09		05-09	92		93-11		
IT02	MONTICOLOR-MONTIGGL	77-13	93	93-14		93-13	93-13	93-10	93-13		-	-	93-01	00	-	-	92-13				92		93-11		
IT03	PASSO LAVAZE	92-08	93-13	92-13		94-13	94-00	93-95	95-07		01-13	-	93-05	94	-	-	93-09	95-09		99-09	92				
IT05	SELVA PIANA	97-08	97-15	97-17		97-17	97-17	95	02-08		-	-	97-05		-	-	97-09	09		99-09					
IT06	PIANO LIMINA	99-08	97-16	97-17		97-17	97-17	95			-	-	97-05		-	-	97-09	09		99-09					
IT07	CARREGA	97-08	97-16	97-17		97-17	97-00	95			98-13	-	97-05		-	-	97-09	09		99-09					
IT09	MONTE RUFENO	97-08	97-16	97-17		97-17	97-00	95	02-08		97-14	-	97-05		-	-	97-09	09		99-09					
IT10	VAL MASINO	97-08	00-15	97-15		97-15		95	05-07		-	-	97-05		-	-	97-09	09		99-09					
IT12	COLOGNOLE	97-01	97-15	97-15		97-15	97-00	95			-	-	97-05		-	-	97-09	09		99-09					
IT13	LA THUILE	97-08	97-15	09-15		09-15		95			-	-	97-05		-	-	97-09			99-08					
LT01	AUKSTAITIJA	93-13	93-17	93-17	93-10	93-17		93-05	94-12	93-17	93-17		06-17	99-17	12		00-17	93-17		02-15	93-17	93-17			93
LT03	ZEMAITIJA	90-13	95-17	95-17	06-10	95-17		94-05	95-12	95-17	95-17		06-17	99-17	95-12		00-17	94-17		02-15	94-17	94-17			94
NO01	BIRKENES	87-17	87-17	87-17	92	89-17		87-11	86-17	87-88	87-17	-	86-17	87-02		-	91-03	86-13			86				
NO02	KÄRVATN	87-91	87-17	87-17	88	89-11		89-13	89-10		87-17	-	89-09	89-02		-	92-03	89-09							
NO03	LANGTJERN		87-97	77-17		86-03		91-13	91-03		87-17		86-03	87-02											
PL01	PUSZCZA BORECKA	06-17	16-17	16-17		16-17		17	10-17		16-17			06-17				16							
PL05	WIGRY	06-17	16-17	16-17		16-17			06-17		16-17			05-17				16							
PL06	PARSENTA	10-17	16-17	94-17		96-17			10-17		94-17			10-17											
PL07	POJEZIERZE CHELMINSKIE	16-17	16-17	16-17							16-17														
PL08	KAMPINOS	09-17	16-17	16-17		16-17		16	12-17		16-17			10-17				16							
PL09	LYSOGORY	05-17	16-17	16-17		16-17			05-17		16-17			05-17				16							
PL10	BESKIDY	94-17	16-17	94-17		02-17			11-17		94-17			09-17				16							
PL11	WOLIN	16-17	16-17	16-17		17			16-17		16-17			16-17											
PL12	ROZTOCZE	16-17	16-17	16-17		16-17			16-17		16-17			16-17				16							
RU03	CAUCASUS BR	89-94	89-17	89-98																					
RU04	OKA-TERRACE BR	89-06	89-17	89-98	90										93-99		93-14	93-02		93		94-96			
RU12	ASTRAKHAN BR	93-94	93-17	93-94																					
RU13	CENTRAL FOREST BR	93	93-94	93													09-17								
RU14	VORONEZH BR	94	94-17	94-98																					
RU16	VELIKIY ISLAND				89-90			89	89	89						93-99	93-17	91-94			89-94	93	94-95		91
SE04	GÄRDSJÖN F1	87-17	88-17	87-17	95	87-16		95-10	87-17	79-17	87-17	-	99-17	96-17			97-01	95-16	91-15	91-15	96-16	92-17	95-17		
SE14	ANEBODA	96-17	96-17	96-17	95	96-16		96-11	95-17	96-17	96-17	-	99-17	95-17			97-01	82-16	96-16	06-16	97-17	97-16	95-17		
SE15	KINDLA	97-17	96-17	96-17		96-16		97-12	95-17	97-17	96-17	-	97-17	95-17			98-01	96-17	98-13	98-13	98-13	97-17	95-17		
SE16	GAMMTRATTEN	99-17	99-17	99-17		99-16		00-18	00-17	00-16	99-17		99-17	00-17			00-01	99-16	99-14	99-14	00-15	00-17	00-17		
UA01	KARADAG	12-13	12-13																						



Figure I.1. Geographical location of ICP IM sites.

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2 Aluminium fractions in surface waters draining catchments of ICP Integrated Monitoring network

Interim report

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2.1 Introduction

Aluminium (Al) is usually unavailable for biogeochemical reactions in the environment because it is largely associated with aluminosilicate minerals. Elevated inputs of strong acids from the anthropogenic atmospheric deposition to sites with low pools of base cations often result in increased concentrations of Al in soil solutions and surface waters. Such situation may be toxic to biota, e.g. aquatic organisms (Gensemer & Playle 1999). Different fractions of aqueous Al have different toxicity levels for aquatic biota, inorganic monomeric Al (Ali, sometimes called labile Al) is generally considered as potentially toxic (Driscoll & Schecher 1990). Several site-specific or regional studies have been performed that consider long-term changes in Al fractions (Palmer & Driscoll 2002, Warby et al. 2008, Krám et al. 2009, Löfgren et al. 2010, McLeod 2016, Driscoll et al. 2016, Riscassi et al. 2018). The objective of this contribution was (1) to collect all and present recently available information about Al fractions from the Integrated Monitoring (IM) database, (2) to stimulate the IM National Focal Points to checkout and add not yet reported Al fractions data to the IM database in SYKE Helsinki, (3) to prepare a scientific paper about Al fractions for the 29th IM Annual Report 2020 and also for an international scientific journal.

2.2 Methods

Modified methods of the original Al fractionation procedure of Driscoll (1984) were applied at some IM catchments (e.g. Røgeberg & Henriksen 1985, LaZerte et al. 1988, McAvoy et al. 1992). Total monomeric Al (Alm) and organic monomeric Al (Alo, sometimes called non-labile Al) were measured in surface water by a colorimetry method. The Alo was separated using a strong cation exchange resin, the method utilizes charge exclusion by ion exchange. Potentially toxic inorganic monomeric Al (Ali) is calculated as the difference between Alm and Alo. Total (dissolved)

aluminum (Al) in surface water was measured using standard methods by Atomic Absorption Spectrophotometry (AAS) or Inductively Coupled Plasma Spectrophotometry (ICP).

2.3 Site description

The ICP IM database contains relevant data about Al fractions in surface runoff from 14 catchments (Fig. 2.1, Table 2.1) for very different time periods (Table 2.2). These catchments belong to seven countries: Czech Republic (1), Finland (5), Estonia (1), Norway (3), Sweden (1), Switzerland (1) and UK (2). The most frequent site conditions contain tills on igneous or metamorphic bedrocks (usually felsic) in morainic landscapes (11), two catchments are on sedimentary bedrocks (Saarejärve, Allt'a Mharcaidh),



June 2019

Figure 2.1. Location of the fourteen ICP IM catchments with reported Al fractions.

and only the Czech catchment does not contain any glacial sediments. Almost all study catchments are forested (Table 2.1). Most of the catchments are situated in low altitudes, three sites have the maximum altitude between 500 and 1000 m a.s.l., two between 1000 and 1500 m a.s.l. and only the Swiss catchment has the maximum altitude almost 3000 m a.s.l. Catchment area varies almost four orders of magnitude.

In total 197 years of data with Al fractions are available for analyses from the period 1987–2017 in the IM database (Table 2.2, status in June 2019). The three Norwegian catchments have the richest database with respect to the Al fractions (29–31 years of data). Considerably long data series are also available from the two British catchments (21 years), but unfortunately not from the last nine years. Valkea-Kotinen has 17 years available, but not from the last eleven years and Lysina has 16 years available in the IM database so far. Twelve years of data are available from Gårdsjön, but with large void between 1996 and 2014, three remaining Swedish catchments did not submit any Al fractions data to the IM database so far in spite that they reportedly analyzed them.

Table 2.1. Basic information about the selected IM catchments (Vuorenmaa et al. 2018, Bergström et al. 1995, Bruder et al. 2016, Söderman 1990, IM database).

Site name ICP IM	Code ICP IM	Area km ²	Altitude m a.s.l.	Predominant vegetation	Predominant bedrocks
Afon Hafren	GB02	3.6	355–690	Sitka and Norway spruce	Mudstone, shale
Allt'a Mharcaidh	GB01	100.0	245–1111	Calluna heath	Granite
Birkenes	NO01	0.4	200–300	Norway spruce, Scots pine	Granite
Gårdsjön	SE04	0.04	114–140	Norway spruce	Granite
Hietajärvi	FI03	4.6	165–214	Scots pine	Granodiorite
Kårvatn	NO02	25.0	200–1375	Scots pine, alpine birch	Gneiss
Lago Nero	CH02	0.8	2385–2842	Grass	Gneiss
Langtjern	NO03	4.8	500–710	Scots pine	Granite, gneiss
Lysina	CZ02	0.3	829–949	Norway spruce	Granite
Musta-Kotinen	FI02	1.6	153–180	Norway spruce, Scots pine	Gneiss
Pesosjärvi	FI04	6.3	256–300	Norway spruce	Greenstone
Saarejärve	EE02	3.3	44–77	Norway spruce, Scots pine	Sandstone
Valkea-Kotinen	FI01	0.3	150–190	Norway spruce, Scots pine	Gneiss
Vuoskojärvi	FI05	1.8	135–240	Mountain birch, Scots pine	Granulite, gneiss

2.4 Results and discussion

Distinct Al patterns were evident in runoff waters of the evaluated IM catchments (Table 2.2). The highest Ali values were detected at Lysina in the Czech Republic (mean 450 µg L⁻¹, median 340 µg L⁻¹) and at Gårdsjön in Sweden (mean 320 µg L⁻¹, median 210 µg L⁻¹). Very high Ali concentrations were measured also at two Norwegian catchments, Birkenes (mean 200 µg L⁻¹, median 170 µg L⁻¹) and Langtjern (mean and median 130 µg L⁻¹). Slightly elevated Ali values were documented at British Afon Hafren (mean 80 µg L⁻¹, median 60 µg L⁻¹) and Estonian Saarejärve (mean 50 µg L⁻¹, median 40 µg L⁻¹). Two catchments situated nearby in southern Finland (Valkea-Kotinen and Musta-Kotinen) have similar values (20–40 µg L⁻¹). The remaining six IM catchments showed very low Ali concentrations in runoff water.

Very high portion (70%) of the total dissolved Al was in the form of Ali at Saarejärve according to data from Table 2.2. More than 40% of Al was presented as Ali at four catchments (Afon Hafren, Kårvatn, Lysina, and Birkenes) and just slightly below 40% of Al was found as Ali fraction at Gårdsjön. Contrary, very low relative proportion

of Ali in Al (<10%) was found at Valkea-Kotinen and Vuoskojärvi in Finland and especially at Scottish Allt'a Mharcaidh.

These presented results are only preliminary. All reported Al results should be taken with caution especially because some sites have shorter dataset for the Ali fraction than for the total Al. Therefore complex and direct comparison of the same time periods will be performed in the next version of this paper, after appropriate communication with the individual data owners and after additions of so far missing Ali values to the IM database.

Table 2.2. Concentrations of inorganic monomeric aluminium (Ali) and total dissolved Al in surface water runoff from the ICP IM network.

Site name ICP IM	Code ICP IM	Number of years	Time span of years	Ali ($\mu\text{g L}^{-1}$)		Al ($\mu\text{g L}^{-1}$)	
				mean	median	mean	median
Afon Hafren	GB02	21	1988–2008	82	60	166	144
Allt'a Mharcaidh	GB01	21	1988–2008	5	2	61	50
Birkenes	NO01	29	1987–2017	201	165	461	457
Gårdsjön	SE04	12	1987–2017	322	208	743	720
Hietajärvi	FI03	5	1992–2006	6	5	30	25
Kårvatn	NO02	29	1987–2017	9	9	20	20
Lago Nero	CH02	2	2016–2017	9	6	5	5
Langtjern	NO03	31	1987–2017	132	128	nd	nd
Lysina	CZ02	16	1991–2017	445	340	951	960
Musta-Kotinen	FI02	8	1989–1998	37	38	283	270
Pesosjärvi	FI04	2	1992–1997	6	6	32	20
Saarejärve	EE02	3	1996–1998	53	43	75	60
Valkea-Kotinen	FI01	17	1989–2006	17	16	189	180
Vuoskojärvi	FI05	1	1993	5	5	68	59

Toxic limits for fish (e.g. $> 50 \mu\text{g L}^{-1}$, Gensemer & Playle 1999) and for benthic macroinvertebrates (e.g. above concentration interval $100\text{--}300 \mu\text{g L}^{-1}$, Herrmann 2001) were reported. Mayfly (*Ephemeroptera*) nymphs are reportedly the most sensitive to Ali from benthic macroinvertebrates (Herrmann 2001). Benthic macroinvertebrates biodiversity at Lysina was low and indeed, mayflies were absent from the stream at Lysina but they were frequently present at other Czech catchments with lower concentrations of Ali and higher streamwater pH values (Traister et al. 2013). Also mean and median concentration of Ali at three other acidic catchments (Gårdsjön, Birkenes and Langtjern) indicates toxic levels for benthic macroinvertebrates and at two more streams concentration indicates problems for fish (Afon Hafren and Saarejärve). Other evaluated IM catchments are probably negatively influenced by elevated concentrations of Ali only during short term high-flow acidic events, especially during spring snow melt.

Time series of total Al (1990–2018) and partially incomplete time series of three Al fractions are available for Lysina (Fig. 2.2). Note the Alp represents the particulate Al fraction calculated as the difference between Al and Alm. Marked decline of Ali concentrations in the 1990s at Lysina coincided with very large decrease of streamwater sulfate and consequent acidification recovery of the catchment. The Ali prevailed as the major Al fraction until the middle of the 2000s. The following decade until the middle of the 2010s exhibited Alp as the major Al fraction. The last three years showed another shift, the Alo as the prevailing Al fraction at Lysina.

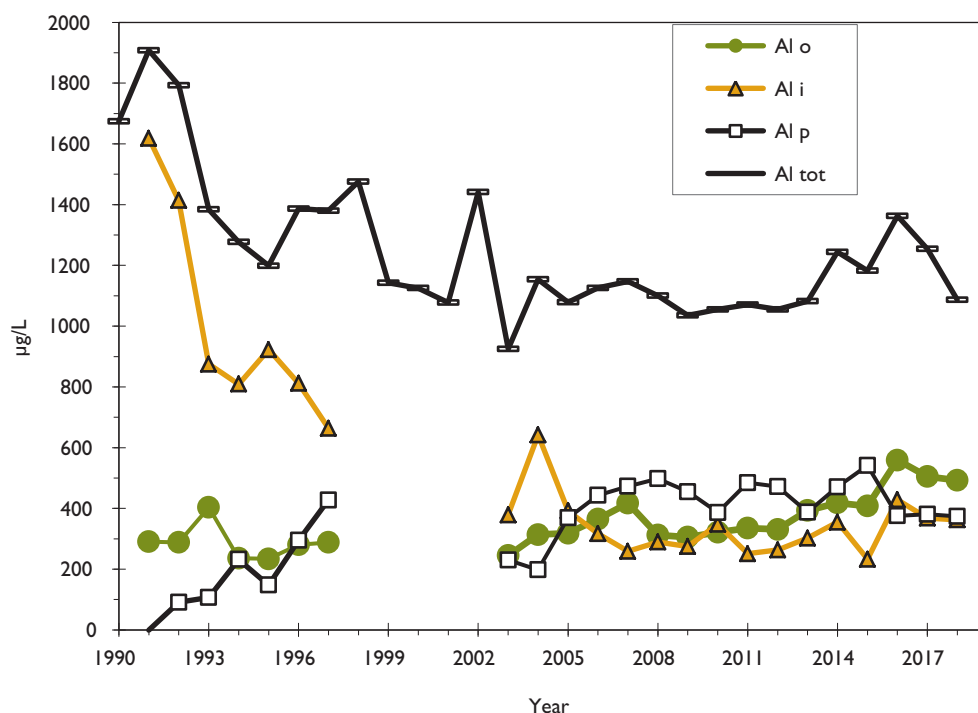


Figure 2.2. Annual discharge-weighted mean streamwater concentrations of Al fractions at Lysina CZ02, calculated for water years (Nov. – Oct.). Ali = inorganic monomeric Al (labile Al), Alo = organic monomeric Al (non-labile Al), Alp = particulate Al, Altot = total Al.

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Annex I

Report on National ICP IM activities in Germany

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Introduction

The German International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) was launched in 1990 at the Forellenbach site (DE01) and in 1998 at Neuglobsow - Lake Stechlin site (DE02). The sites are far away from the main sources of air pollutants and subject to very similar changes in global change characteristics (composition and rates of atmospheric deposition, warming) despite greatly differing concerning physiographic and environmental conditions (Table 1).

DE01 in the Bavarian Forest National Park, the second highest low mountain range in Germany has low air temperature (6.5°C) and high precipitation (1536 mm) with considerable variation due to altitudinal gradients (787-1292 m a.s.l.). Bedrock is granite and gneiss, soils are nutrient-poor throughout (mostly dystic cambisols) and wet on 30% of catchment area (69 ha). Norway spruce covers cold and wet sites while European beech dominates mixed forests growing on warmer slopes. From 1992 to 2007, forests experienced two bark beetle (*Ips typographus* L.) outbreaks, killing mature spruce stands on 60% of the area. Therefore, regenerating spruce and mixed forests are the prominent vegetation units.

Table 1. Catchment characteristics and long-term means (range) of air temperature and key figures of the hydrological cycle at German IM sites.

	DE01 Forellenbach (1992–2018)	DE02 Neuglobsow (1998–2017)
Latitude, Longitude	48°57' N, 13°25' E	53°08' N, 13°02' E
Altitude (MASL)	899 (787–1292)	65
Area (ha)	69	1420
Bedrock	Granite, gneiss	Weichselian glacial sand
Soils	70% lithosol – dystic (podsol) cambisol, 30% histosol, gleysol	Haplic arenosol
Vegetation	70% Norway spruce, 30% broadleaves, mostly European beech	20% Scots pines, 80% European beech
Temperature (°C)	6.5 (4.9–7.7)	9.3 (7.9–10.1)
Precipitation (mm)	1536 (1006–2209)	600 (422–816)
Runoff (mm)	988 (699–1449)	84 (2–257) seepage
Evapotranspiration (mm)	548	516

The Neuglobsow site (DE02) located in the north-eastern German lowlands is characterised by mixed forests with 160-year-old Scots pines and 110-year-old beech trees undisturbed since 1938. The subcontinental climate has a mean annual air

temperature of 7.9°C and a mean annual precipitation of 658 mm in the period from 1951 to 1980 (Richter 1997). The soil type according to the World Reference Base for Soil Resources (WRB) is a haplic arenosol with a low capacity of plant available water. The monitoring site is measuring air quality since 1974. It is one of six German EMEP stations and one out of two GAW regional stations and a background station according to the EU air quality legislation. The catchment covers an area of 1420 ha.

Water cycle

Since 1992, annual precipitation declined by 312 mm ($p < 0.01$) at Forellenbach DE01. Two thirds of this decrease in precipitation occurred in the winter half-year affecting groundwater recharge, which depends mostly on winter precipitation. Consequently, the groundwater level at three measuring points in or adjacent to the study area has dropped by 65–86 cm ($p < 0.05$). Alternatively, just as strongly and significantly, it sank after break-points between 2005 and 2007 following a break-point in winter precipitation (2003). The four years with the lowest mean groundwater levels since 1992 were 2014, 2016, 2017 and 2015; 2018 was ranked 8th. Declining winter precipitation contrasts with regional climate change projections but is a possibly still increasing risk for the regional drinking water supply from upper groundwater layers.

Since 1972, mean annual and mean summer air temperature raised by 1.7 K ($p < 0.001$) thereby significantly increasing evapotranspiration rates (Beudert et al. 2018). The year 2018 was the second warmest year since measurements began (7.6°C) and sunshine duration reached a new record value (1830 hours).

Despite decreasing precipitation and increasing evapotranspiration trends, catchment outflow didn't react adequately. Rapidly growing stand regeneration uses less water than mature stands. So bark beetle effects on 60% of catchment area currently but temporarily compensate for climate change effects (Bernsteinová et al. 2015).

At ICP Integrated Monitoring site Neuglobsow the water budget of this mature beech and pine stand was evaluated over a 20-year period from 1998 till 2017. The SVAT model Expert-N was used to predict soil water storage and water fluxes, i.e. evapotranspiration, interception and groundwater recharge.

The results show already existing negative trends of water components during this period. Average annual precipitation was 600 mm which corresponds to a minus of 9% compared to the long-term mean of Richter (1997) in the period 1951–1980. Average annual temperature in the last two decades was 9.3°C which represents an increase of 1.4°C (Fig.1).

Potential transpiration has thus increased by 10% and groundwater recharge has dropped sharply by 12% in the last decade compared to the long-term annual average (Nützmann 2003). In contrast, actual transpiration rates remained unchanged during these years. Water stress indicators such as RTI (relative transpiration index) and REW (relative extractable water) also showed a significant increase of days with water stress especially during the growing season (Schulte-Bisping & Beese 2013).

Sulphur and nitrogen deposition

The sulphur deposition decreased to $\leq 2 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in 2018, a reduction by 75–90% compared to the early 1990s but most of it occurred up until 2000. At Neuglobsow site, S deposition decreased to $\leq 3 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in 2017, while at the beginning of the investigations in 1998 S deposition was more than $5 \text{ kg ha}^{-1} \text{ yr}^{-1}$. The strongest reduction has already happened before or around the year 1990 when air SO_2 concentrations were 10 times higher than today. This indicates that atmospheric acidification by S deposition no longer poses a major risk to the vitality and stability of forest ecosystems on these two sites.

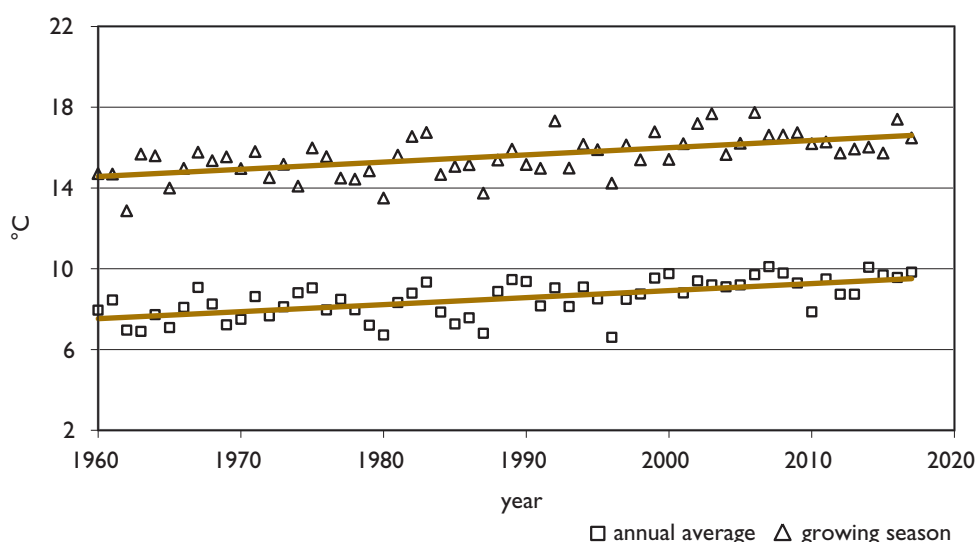


Figure 1. Long-term means of air temperature at Neuglobsow DE02 (1960–2017).

The long-term deposition rates of inorganic nitrogen DIN show only very small differences between open site and forest stands at Forellenbach DE01 concerning both, mean ($9\text{--}10\text{ kg ha}^{-1}\text{ yr}^{-1}$) and standard deviation ($\sim \pm 2\text{ kg ha}^{-1}\text{ yr}^{-1}$) (Table 2). The 2018 value of spruce throughfall DIN was similar to the long-term mean. By contrast, beech throughfall and bulk DIN deposition were below average and time series show a reduction of 3.2 ($p < 0.01$) and $5.1\text{ kg ha}^{-1}\text{ yr}^{-1}$ ($p < 0.001$) since 1992. In 2018, bulk and wet-only DIN deposition offered the same rate of $5.9\text{ kg ha}^{-1}\text{ yr}^{-1}$ indicating the small relevance of particle deposition at this site.

Table 2. Throughfall and bulk deposition ($\text{kg ha}^{-1}\text{ yr}^{-1}$) of dissolved inorganic nitrogen (DIN) at German IM sites.

	DE01 1992–2017	DE01 2018	DE02 1998–2017	DE02 2018
wet-only		5.9		
bulk	9.6 ± 2.0	5.9	6.0 ± 0.5	4.6
throughfall beech	9.2 ± 1.8	7.2		
throughfall spruce	9.8 ± 1.8	9.2		
throughfall mixed beech pine			6.4 ± 0.6	5.9

Total DIN deposition estimate (2003–2018) was calculated from bulk deposition and throughfall DIN (Draijers & Erisman 1995) by using canopy budget models. Adding the difference between throughfall and bulk dissolved organic nitrogen DON (ΔDON) revealed the total deposition of reactive nitrogen Nr (assuming that ΔDON results from conversion of air-borne DIN by microorganisms). From 2003 to 2018, total reactive nitrogen deposition Nr was 11.3 ± 2.0 and $15.1 \pm 2.2\text{ kg ha}^{-1}\text{ yr}^{-1}$ in beech and spruce stands respectively.

By combining mutually supporting measurement methods for the total reactive nitrogen and/or particulate components (eddy-covariance, denuder, passive collectors, gas analyzers) with different modelling approaches, concentrations and fluxes of N_r at DE01 measuring tower could be determined for the years 2016 and 2017 (FORESTFLUX, UBA Fkz. 3715512110, Brümmer et al. 2019). Dry deposition rates weighted by tree species were $4.4\text{ kg N ha}^{-1}\text{ yr}^{-1}$ for the eddy-covariance approach and 5.2 to $6.9\text{ kg N ha}^{-1}\text{ yr}^{-1}$ for modelling with DEPAC-1D and DEPAC in LOTOS-EUROS.

Dry deposition rates from canopy budget methods span the same value range (3.9 and 6.5 kg N ha⁻¹ yr⁻¹) of ecophysiologicaly consistent results. The main component in the dry deposition was reduced nitrogen (76%), followed by HNO₃ (11%), NO₂ and NO (Zöll et al. 2019).

At Neuglobsow site, long-term bulk nitrogen deposition (1998–2017) in form of ammonium and nitrate was 3.1±0.6 and 2.9±0.4 kg N ha⁻¹ yr⁻¹, respectively (in total 6.0 ± 0.5 kg N ha⁻¹ yr⁻¹). Throughfall DIN resulted in deposition rates of 3.1±0.6 N_r and 3.3±0.6 N_o ha⁻¹ yr⁻¹. Meanwhile, the average gaseous loss in form of N₂O was 0.5 kg N ha⁻¹ yr⁻¹ and the leaching loss was 2.1 kg N ha⁻¹ yr⁻¹.

Total DIN deposition estimate was calculated from bulk deposition and throughfall DIN by using the canopy budget model of Ulrich (1983). From 1998 to 2017, average total nitrogen deposition was 13.0±2.0 kg ha⁻¹ yr⁻¹.

Conclusions

The results highlighted in this report represent only an extract of the collected and modelled data on both the German Integrated Monitoring sites. However, it already offers important insights of trends for main pollutants (e.g. nitrogen and sulphur deposition) and the development of ecologically relevant climatic parameters. Especially the integration of measurements and modelling results along the whole catchment (e.g. precipitation, evapotranspiration and outflow) provides a deeper understanding of complex processes within the ecosystem.

Acknowledgments

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Annex 2

Report on National ICP IM activities in Sweden in 2017

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Introduction

The Swedish integrated monitoring programme is run on four sites distributed from south central Sweden (SE14 Aneboda), over the middle part (SE15 Kindla), to a northerly site (SE16 Gammtratten). The long-term monitoring site SE04 Gårdsjön F1 is complementary on the inland of the West Coast and has been influenced by long-term high deposition loads. The sites are well-defined catchments with mainly coniferous forest stands dominated by bilberry spruce forests on glacial till deposited above the highest coastline. Hence, there has been no water sorting of the soil material. Both climate and deposition gradients coincide with the distribution of the sites from south to north (Table 1). The forest stands are mainly over 100 years old and at least three of them have several hundred years of natural continuity. Until the 1950's, the woodlands were lightly grazed in restricted areas. In early 2005, a heavy storm struck the IM site SE14 Aneboda. Compared with other forests in the region, however, this site managed rather well and roughly 20–30% of the trees in the area were storm-felled. In 1996, the total number of large woody debris in the form of logs was 317 in the surveyed plots, which decreased to 257 in 2001. In 2006, after the storm, the number of logs increased to 433, corresponding to 2711 logs in the whole catchment. In later years, 2007–2010, bark beetle (*Ips typographus*) infestation has almost totally erased the old spruce trees. In 2011 more than 80% of the trees with a breast height over 35 cm were dead (Löfgren et al. 2014) and currently almost all spruce trees with diameter of ≥ 20 cm are dead.

Table 1. Geographic location and long-term climate and hydrology at the Swedish IM sites (long-term average values, 1961–1990).

	SE04	SE14	SE15	SE16
Latitude; Longitude	N 58° 03'; E 12° 01'	N 57° 05'; E 14° 32'	N 59° 45'; E 14° 54'	N 63° 51'; E 18° 06'
Altitude, m	114–140	210–240	312–415	410–545
Area, ha	3.7	18.9	20.4	45
Mean annual temperature, °C	+6.7	+5.8	+4.2	+1.2
Mean annual precipitation, mm	1000	750	900	750
Mean annual evapotranspiration, mm	480	470	450	370
Mean annual runoff, mm	520	280	450	380

In the following, presentation of climate, hydrology, water chemistry and some ongoing work at the four Swedish IM sites relate mainly to the year 2017 (Löfgren 2018).

Climate and Hydrology in 2017

In 2017, the annual mean temperatures were higher (0.6–1.1 °C) compared to the long-term mean (1961–1990) for all four sites. Largest deviation occurred at the northern SE16 Gammtratten site. Compared with the measured time series, 17 years at site SE16 Gammtratten and 21 years at the other sites, the temperatures in 2017 were somewhat higher at the two southern IM sites (0.5 and 0.7 °C) while the two northern sites actually showed lower values with 0.3 and 0.4 °C. The annual mean values were slightly lower compared to the period 2014–2016 when temperatures were the highest observed for the whole measurement period with exception for SE15 Kindla where the temperature was slightly higher in the years 1999 and 2000. The variations between years have been considerable, especially for the last five years, over 3° C at three of the sites. Smaller variations were found at the central site SE15 Kindla, only 1 °C. Low temperatures were observed in the years 2010 and 2012 with 3.1–3.6 °C below the 21 year mean at three sites while SE15 Kindla only deviated with 1.3 °C below.

Compared to the long-term average values (1961–1990), the precipitation amounts in 2017 were close to average at SE14 Aneboda and SE15 Kindla (2 and 6% excess). For SE04 Gårdsjön 20% higher precipitation was observed. Only SE16 Gammtratten in the north followed previous year with lower precipitation than the long-term mean, reaching 83%. This was similar to 2016. For site SE04 Gårdsjön, the precipitation amount compared to average was comparably low in May and July while most other months had higher values.

The characteristic annual hydrological patterns of the catchments are for the southern sites high groundwater levels during winter and lower levels in summer and early autumn. In northern locations, water levels often are low in winter when precipitation is stored as snow, raising levels at snowmelt in spring and turning to lower levels in summer due to evapotranspiration. However, depending on rainfall amounts in summer, the groundwater levels could occasionally be elevated also in this period. Rainfall in autumn would yield the same result. In 2017 at SE14 Aneboda, slightly elevated groundwater levels occurred in spring and also in June due to high rainfall (129 mm). Autumn was quite wet, starting already in August with consecutive higher and higher levels until the end of the year. In the central parts of the catchment, the groundwater pressure got artesian. For SE16 Gammtratten in the north, snowmelt occurred in May and rather high rainfall in June resulted in high groundwater levels in June. After that, the groundwater level was subsiding, but 160 mm precipitation in September–October elevated the levels again. Later, cold weather and snow made the groundwater levels to recede. At site SE15 Kindla, a more varying pattern was observed with several peaks 0.2 m below the soil surface during snowmelt in March–April, summer rains in June and also in autumn created groundwater level peaks. The lowest levels, 0.8 m below soil surface, were observed in early August whereafter rain successively elevated the groundwater levels until the end of the year. These patterns were fairly similar to those in 2015. The groundwater levels were reflected in the stream water discharge patterns (Fig. 1).

In addition to precipitation, evapotranspiration affects the runoff pattern. The runoff pattern for SE16 Gammtratten, was fairly typical but with a snowmelt peak in May and a higher discharge in October. At SE04 Gårdsjön, the pattern was in accordance with the average except for at the end of the year when runoff was higher than normal in December. Runoff at SE15 Kindla followed the ordinary pattern during the first half of the year where after it subsided and was low until October. Thereafter runoff increased to higher values during the last two months of the year. Runoff at

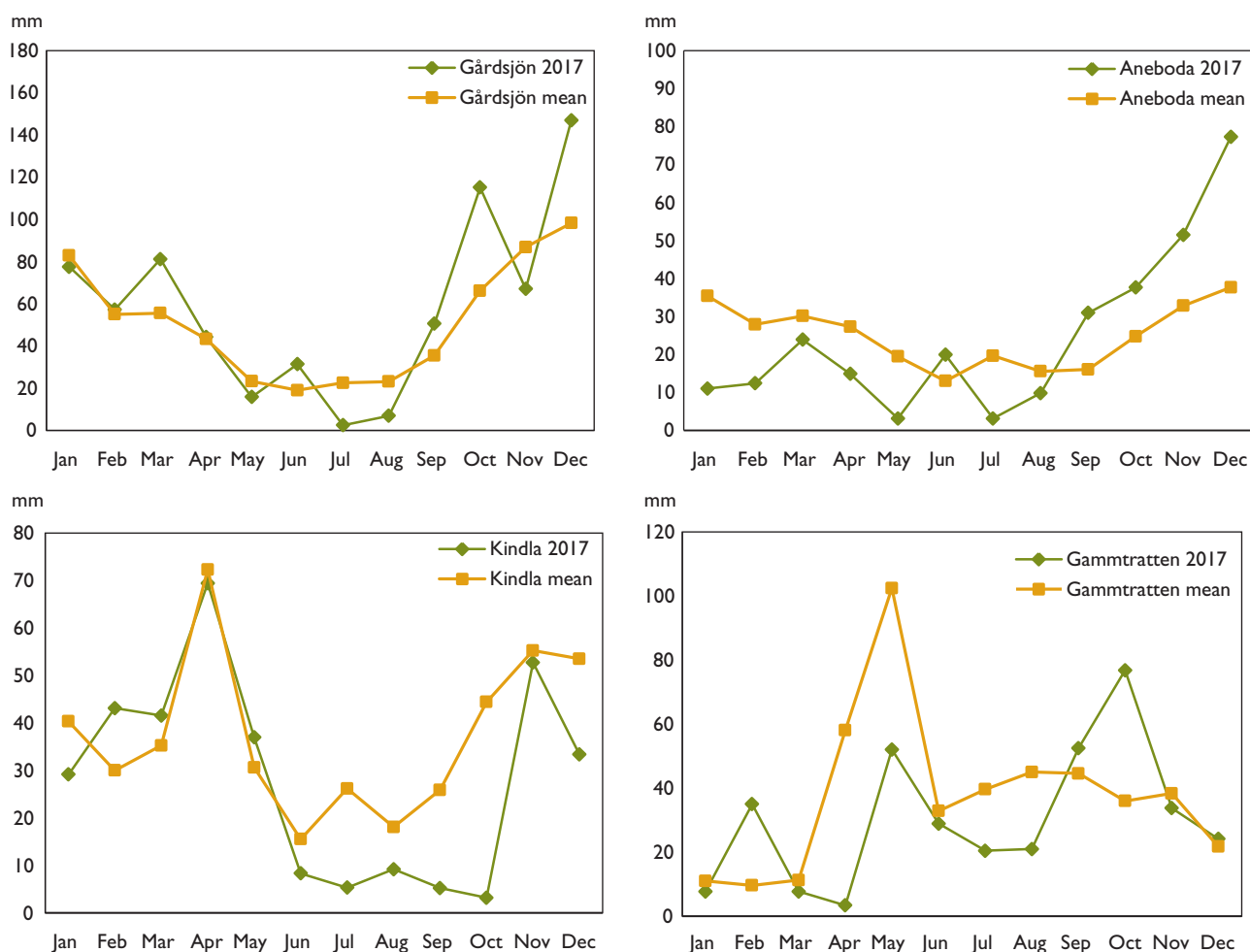


Figure 1. Discharge patterns at the Swedish IM sites in 2017 compared to monthly averages for the period 1996–2017 (mean). Note the different scales at the Y-axis.

SE14 Aneboda showed slightly lower monthly values in the beginning of the year, turning high during the last four months (Fig. 1) in line with the groundwater levels.

At the two northern sites, generally, snow accumulates during winter, resulting in low groundwater levels and low stream water discharge. However, warm winter periods with temperatures above 0 °C have during a number of years contributed to snowmelt and excess runoff also during this season. Consequently, the spring discharges have been comparably low during snowmelt, deviating from the normal conditions, this could be seen at SE16 Gammtratten. In southern Sweden, SE04 Gårdsjön and SE14 Aneboda the situation deviated somewhat from the average pattern with higher runoff than normal in autumn (Fig. 1).

In 2017, the annual runoff made up 27–63% of the annual precipitation (Table 2), a wide range compared to the ordinary 40–60% found in previous years except for 2016 when the range was even larger (31–83%). The highest share was found at the southwest site SE04 Gårdsjön (63%), due to high runoff in the end of the year when evapotranspiration was low (Table 2). Runoff at this site, being almost 2/3 of precipitation would be quite normal. At SE14 Aneboda, storm felling, followed by bark beetle attacks, have reduced the forest canopy cover, inducing low interception. Actually, the measured throughfall reached 94% of the precipitation (89% in 2016). The total evapotranspiration was estimated to 477 mm (349 mm in 2016), a value considerably higher than in the previous years. At SE15 Kindla, the water balance was rather normal, however, with slightly high evapotranspiration and somewhat low runoff. At

the northern site SE16 Gammtratten, throughfall and bulk precipitation were very similar (1% deviation), which is erroneous and indicates large uncertainties in any of these two measurements. Presumably, snow deposition in bulk precipitation infers the largest uncertainty.

Table 2. Compilation of the 2017 water balances for the four Swedish IM sites. P – Precipitation, TF – Throughfall, I – Interception, R – Water runoff

	Gårdsjön SE04		Aneboda SE14		Kindla SE15		Gammtratten SE16	
	mm	% of P	mm	% of P	mm	% of P	mm	% of P
Bulk precipitation, P	1112	100	772	100	977	100	624	100
Throughfall, TF	909	82	729	94	595	61	630	101
Interception, P–TF	203	18	44	6	382	39	-6	-1
Runoff, R	696	63	295	27	415	42	363	58
P–R	416	37	447	73	562	58	261	42

Water chemistry in 2017

Low ion concentrations in bulk deposition (electrolytical conductivity 1–2 mS m⁻¹) characterise all four Swedish IM sites. The concentrations of ions in throughfall, including dry deposition, were higher at the three most southern sites. At the northern site SE16 Gammtratten, the conductivity in throughfall (0.7 mS m⁻¹) was almost the same as in bulk deposition indicating very low sea salt deposition and uptake of ions by the trees. At the two most southern sites, sea salt deposition provides tangibly higher ion concentrations, especially at the west coast SE04 Gårdsjön site (4.9 mS m⁻¹ in throughfall).

The groundwater pathways are fairly short and shallow in the catchments, providing rapid soil solution flow paths from infiltration to surface water runoff. However, the conductivity in soil water was higher compared to throughfall showing influences from evapotranspiration and soil chemical processes. The deposition acidity has during the last 10 years been rather similar at all sites with somewhat higher pH values (0–0.5 units) in throughfall compared with bulk deposition. However, in 2017, SE04 Gårdsjön had a throughfall pH 5.2 while the two sites SE14 Aneboda and SE15 Kindla had values c. 5.4 (Table 3). For SE16 Gammtratten, the pH value was 5.2 both in bulk deposition and in throughfall.

Table 3. Mean deposition chemistry values 2017 at the four Swedish IM sites. S and N in kg ha⁻¹ yr⁻¹.

	SE04	SE14	SE15	SE16
pH, bulk deposition	5.1	5.1	5.4	5.2
pH, throughfall	5.2	5.5	5.4	5.2
S, bulk deposition	3.3	1.3	1.4	0.8
N, bulk deposition	8.6	4.0	4.9	1.7

During the water passage through the catchment soils, organic acids were added and leached to the stream runoff. In the upslope recharge areas, pH in the upper soil layers (E-horizon) was mainly lower than in throughfall. However, in the peat in discharge areas at SE15 Kindla and SE16 Gammtratten, pH was higher compared to throughfall while it was slightly lower compared to throughfall at SE14 Aneboda but considerably lower at SE04 Gårdsjön with a pH of 4.3. In the recharge areas, the buffering capacity in soil water and groundwater varied between negative and positive values, but was most frequently on the negative side, especially for SE04 Gårdsjön

with constantly negative values. In the discharge areas, the buffering capacity in groundwater was fairly high with ANC exceeding 0.22 mEq L^{-1} at SE14 Aneboda and SE15 Kindla and with bicarbonate (HCO_3^-) occasionally present at Aneboda, Kindla and Gammtratten at average concentrations of 0.02, 0.14 and 0.05 mEq L^{-1} , respectively. At SE04 Gårdsjön ANC was negative (-0.01 mEq L^{-1}). The stream waters were acidic with pH values below 4.7 at all sites except Gammtratten having a pH of 5.6. The stream water buffer capacity was positive at all sites ($\text{ANC} \geq 0.004 \text{ mEq L}^{-1}$), except for SE04 Gårdsjön ($\text{ANC} -0.022 \text{ mEq L}^{-1}$). Anions of weak organic acids and bicarbonate contributed to the positive ANC (0.1 mEq L^{-1}) at SE16 Gammtratten.

The share of major anions in bulk deposition was similar for sulphate, chloride and nitrate at three of the sites, while chloride dominated at SE04 Gårdsjön due to the proximity of the sea. Sea salt showed clear influences on throughfall at SE04 Gårdsjön and also at SE14 Aneboda indicating effects of dry deposition. In throughfall, organic anions contributed significantly at all four sites. The chemical composition changed during the catchment soils passage and the sulphate concentrations were higher in stream water compared with deposition, indicating desorption or mineralization of previously accumulated sulphur in the soils. For Aneboda, nitrification contributed to fairly high nitrate values in the recharge area soil water ($0.02\text{--}0.23 \text{ mEq L}^{-1}$), values being lower compared to previous year. Considerably lower concentrations occurred in the discharge areas, probably due to nitrogen uptake and denitrification.

For site SE16 Gammtratten in the north, sulphate concentrations in soil water and stream water were considerably higher compared to throughfall, indicating release from the soil pool. Organic anions dominated anion flow in the stream with 2/3 of the content to be compared to 25% in SE14 Aneboda and SE15 Kindla reaching only 10% in SE04 Gårdsjön.

Besides effects on ANC and pH, the stream water chemistry is to a considerable extent influenced by organic matter. At SE14 Aneboda, the DOC concentration was high with 28 mg L^{-1} while the other sites SE04 Gårdsjön, SE15 Kindla and SE16 Gammtratten showed lower values 14, 10, and 10 mg L^{-1} , respectively. High DOC concentrations create prerequisites for metal complexation and transport as well as high organic nitrogen fluxes. The organic nitrogen concentrations in stream water ranged from 0.18 to 0.66 mg N L^{-1} . The shares of Norg/Ntot were 87–90%, showing Norg dominating Ntot, and with SE14 Aneboda having the lowest share while SE16 Gammtratten and SE15 Kindla were on the highest range. Inorganic nitrogen ($\text{NO}_3\text{--N}$ and $\text{NH}_4\text{--N}$) was low at the two sites SE15 Kindla and SE16 Gammtratten with 15 and 7 mg L^{-1} , respectively. Somewhat higher concentration in SE04 Gårdsjön with 43 mg L^{-1} reflecting still somewhat high deposition. Higher concentration in stream water was noticed for SE14 Aneboda with 100 mg L^{-1} , possibly due to the forest damage. However, compared to 2016 value 191 mg L^{-1} , the inorganic N concentrations decreased considerably.

Total phosphorus (Ptot) in bulk deposition varied between 5 and $14 \text{ } \mu\text{g L}^{-1}$ with the highest values at SE04 Gårdsjön and lowest in the northernmost site. In stream water, SE14 Aneboda also showed the highest Ptot ($22 \text{ } \mu\text{g L}^{-1}$) as well as DOC concentrations. The other sites had average Ptot concentrations between 3 and $6 \text{ } \mu\text{g L}^{-1}$ with the lowest value at SE15 Kindla.

Inorganic aluminum (Al_i), toxic to fish and other gill-breathing organisms, has been analyzed in soil solution, groundwater and surface waters at the IM sites. Relatively high total Al concentrations occurred in the soil solution ($0.7\text{--}3.6 \text{ mg L}^{-1}$) as well as in stream water ($0.25\text{--}0.50 \text{ mg L}^{-1}$) at the southern sites SE14 Aneboda and SE15 Kindla with low pH (c. 4.8). At the northern site SE16 Gammtratten with a pH of 5.6, the total Al concentrations were low, approximately 0.23 mg L^{-1} and higher in SE14 Aneboda and SE15 Kindla with 0.5 mg L^{-1} . Inorganic Al made up 13–44% of the total Al with the highest value in SE15 Kindla and lowest in SE16 Gammtratten,

corresponding to 0.03–0.22 mg Al_i L⁻¹ with high Al_i at low pH, and the 0.03 mg Al_i L⁻¹ at the northern site SE16 Gammtratten with higher pH. According to the SEPA classification system, the Al_i concentrations at SE04 Gårdsjön, SE14 Aneboda and SE15 Kindla are considered extremely high and high at SE16 Gammtratten. The priority heavy metals Pb, Cd and Hg were still accumulating in the catchment soils, while the stream concentrations were low compared with the levels causing biological effects. However, methyl mercury, only measured at Aneboda and financed by SITES, was still relatively high creating prerequisites for bioaccumulation. In stream water Hg-tot concentration was 8.3 ng L⁻¹ with Hg-methyl on 2.5 ng L⁻¹.

In summary, the four Swedish IM sites show low ion contents and permanently acidic conditions. In stream water, only the northern site SE16 Gammtratten had buffering capacity related to bicarbonate alkalinity. Organic matter has an impact on the water quality with respect to colour, metal complexation, and phosphorus concentrations at all sites, but less at SE15 Kindla, where rapid soil water flow paths provide low DOC and acidic waters. For SE14 Aneboda, the forest dieback provides a relatively high share of water runoff as well as high nitrate concentrations compared with the other three sites. In SE04 Gårdsjön, deposition is strongly influenced by the sea.

Major disturbances test forest resilience

The impact of disturbances on boreal forest plant communities is not fully understood, particularly when different disturbances are combined, and enduring changes in the dominant species are possible after disturbance. Our study site is a long term monitored semi-natural forest in Sweden (SE14 Aneboda) which was subject to intense combined storm and bark beetle damage, beginning with storm Gudrun in 2005. This provided a valuable opportunity to investigate the post-disturbance development of the vegetation community (Weldon & Grandin 2019). Previous studies suggested that a shift from a Norway spruce to a beech dominated forest was possible here, and field workers had remarked on a drastic increase in beech saplings.

We analysed pre- and post-disturbance vegetation data to investigate to what extent vascular plant species abundances, diversity, traits, and community composition have changed. We were particularly interested in differences between the remaining apparently unaffected areas (which could potentially act as refuges) and disturbed areas, and in signs of consistent change over time in community composition in response to disturbance that could indicate an impending regime shift (to a beech dominated state for example).

We found that the vegetation community present in the refuge areas has remained substantially intact throughout the period of disturbance. However, non-refuge areas diverged over time from the refuges in community composition and showed increased taxonomic and functional diversity. Despite this, an increase in deciduous tree species (particularly beech), spruce has shown strong post-disturbance regeneration across the site. The refuges are likely to be important as a seed source in the apparent ongoing recovery of the disturbed areas to a spruce-dominated state similar to that found pre-disturbance. This fast recovery is evidence of a system resilient to a potential shift to a deciduous-dominated state.

Our results show that even powerful combined disturbances in a system with alternative stable states can be insufficient to initiate a regime shift. The resilience of the spruce-dominated forest community is increased by the survival of refuge areas functioning as a form of ecological memory of the previous ecosystem state. Finally, it is important to note that studies such as this are only possible with the valuable data generated by long-term monitoring programs.

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The Integrated Monitoring Programme (ICP IM) is part of the effect-oriented activities under the 1979 Convention on Long-range Transboundary Air Pollution, which covers the region of the United Nations Economic Commission for Europe (UNECE). The main aim of ICP IM is to provide a framework to observe and understand the complex changes occurring in natural/semi natural ecosystems.

This report summarizes the work carried out by the ICP IM Programme Centre and several collaborating institutes. The emphasis of the report is in the work done during the programme year 2018/2019 including:

- A short summary of previous data assessments
- A status report of the ICP IM activities, content of the IM database and geographical coverage of the monitoring network
- An interim report on aluminium fractions in surface waters draining catchments of ICP Integrated Monitoring network
- National Reports on ICP IM activities are presented as annexes.



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